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A liquid jet sample environment for experiments on liquids at MID (EuXFEL)

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Abstract. An experimental setup for liquid jet applications was implemented at the Materials Imaging and Dynamics Instrument (MID) of the European XFEL. This setup is operated in the multi-purpose experimental chamber of MID at pressures down to 10^{-5} mbar. The motion of the nozzle was realized in three dimensions for alignment purposes and the investigation of liquids at different states of supercooling. For this purpose, an active nozzle temperature control is implemented and a catcher system has been installed to capture the liquid after passing the interaction region to maintain good vacuum conditions. The setup is compatible with different nozzle designs. For spill protection, additional housing with secondary pumping around the nozzle, liquid jet and catcher was implemented. A magnifying camera system for aligning the jet and shadow imaging is available. The setup has already been commissioned and operated with different jets and detector configurations.

1. Introduction

In the last decades, the technological and scientific interest in water and complex liquids has increased. In particular, at synchrotron-radiation and free-electron laser (FEL) sources the development of liquid jets was promoted for such studies [1-3]. Liquid jets offer a container-free, self-refreshing [4] and free-flowing sample system with controlled sample consumption. These conditions are important, especially for radiation and pressure-sensitive biological samples [5,6]. Up to now different types of nozzles have been developed depending on the application [7]. For example, ultra-thin or flat liquid jets are used in infrared and soft X-ray spectroscopy [8], µm-sized liquid droplets were studied at hard X-ray FEL facilities [9] and jets can be used for mixing liquids in reaction studies [7,10]. Injected liquid jets in vacuum facilitate temperature-dependent studies of the liquid making use of evaporative cooling of the liquid with increasing distance from the nozzle [11-13]. This allowed already studying of various conditions and processes, for example, the supercooled state of liquid water where many anomalies occur [14], as well as crystal nucleation of atomic liquids [15].

In this work, the design of a liquid jet setup operated in various detector configurations at the Materials Imaging and Dynamics instrument (MID) at European XFEL (EuXFEL) [16] is presented. It is particularly designed for structural and dynamics studies of aqueous solutions and has been used in X-ray diffraction and split-delay X-ray photon correlation studies. Different nozzle types can be installed, including Rayleigh and gas-dynamics virtual nozzles (GDVN). Using piezo actuators in addition, trains of μm-sized droplets can be generated [17,18].

2. Technical design

Figure 1a shows a photo of the liquid jet setup mounted on an additional hexapod system underneath, inside the multi-purpose vacuum chamber of the MID instrument [16]. The photo was taken through the X-ray exit gate valve. The setup is installed inside the vacuum chamber, but can also be operated in air or gas atmosphere. All motorized motions can be controlled via the beamline operation software Karabo [19]. The temperature can be set remotely. The liquid pumps for running the liquid jet are located outside the vacuum chamber. They are connected with tubing via a feedthrough flange to the nozzle. Typically, for most applications a stack of Shimadzu HPLC pumps is used. Some nozzles, such as GDVN, require a constant flow of focusing gas, e.g. helium, which is provided by a separate gas line allowing a pressure of up to 40 bar. Depending on the jet size a vacuum level of up to 10^{-5} mbar in the multi-purpose vacuum chamber is reachable. For

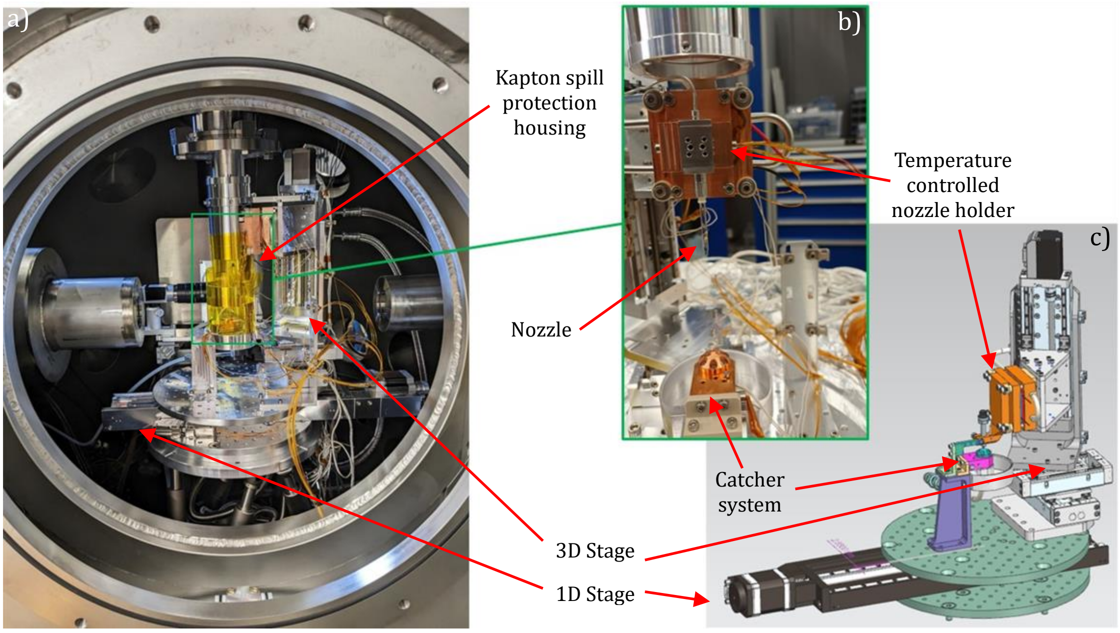

Figure 1. a) Photo of the liquid jet setup mounted on the hexapod inside the multi-purpose vacuum chamber of the MID instrument. The photo was taken through the X-ray exit gate valve where the detector will be installed for LFOV configuration. In the green box, inside an additional Kapton housing, the nozzle and the catcher systems are installed in LFOV configuration. b) Photo of the liquid jet setup before mounting in the multi-purpose chamber of MID instrument. In the top part of the photo the temperature-controlled copper nozzle holder is shown. At the bottom of the photo the top part of the catcher system is visible. c) CAD schematic of the setup. The adapter-plate for the nozzle holder is shown in orange, in this case for an in-house made 3D printed nozzle. The catcher top part is shown in turquoise and the catcher body in purple.

most experiments a vacuum of approximately 10^{-3} mbar has become common for jets of a few micrometres up to 50 μm jet diameter.

X-ray detectors can be set in several configurations. The main detector of the MID instrument is the Adaptive Gain Integrating Pixel Detector (AGIPD) 1M [20,21], it can be configurated in the large-field-of-view (LFOV) configuration with a sample-detector sensor layer distance of down to approximately 17 cm as well as in small-angle X-ray scattering (SAXS) or wide-angle X-ray scattering (WAXS) configuration placing the AGIPD several meters downstream of the interaction point [16]. In WAXS configuration the diffracted X-ray beam exits the multi-purpose chamber and enters the detector fly tube via Kapton windows. In the LFOV and SAXS configurations, due to the windowless connection of the sample chamber and detector and the vicinity of AGIPD to the sample, strict vacuum conditions are needed.

Figure 1b shows the inside of the Kapton housing (green box in Figure 1a) where the nozzle and the catcher system are installed. The Kapton housing can optionally be installed for spill protection. The housing is connected to an additional Turbo pump mounted on the top flange of the sample chamber, to additionally reduce pressure and removing evaporated sample gas. A photo of the liquid jet setup without the housing and before mounting into the multi-purpose chamber of the MID instrument is shown in Figure 1b. The temperature control is realized by pre-regulating a copper heat sink using a water circulation backed to a fine adjustment of the nozzle holder using a Peltier-heating/cooling and a heating element. The copper nozzle holder adapter connecting the Peltier-element and the nozzle allows the installation of several types of nozzle holders. In Figure 1b a quartz-glass Rayleigh nozzle was attached. At the bottom of the photo, the top part of the catcher system is visible. The orifice size, as well as the material of the catcher top can be changed by replacing the blunted cone-like top part. Depending on the jet dimensions and the targeted pressure level this can be a key parameter. Orifice sizes of 300 μm, 500 μm and 1000 μm are available. Figure 1c shows a Computer-Aided Design (CAD) schematic of the setup for a better overview. The nozzle is mounted on a motorized stage that can be moved in three dimensions to align the jet to the catcher's orifices and varying the distance of the nozzle orifice from the focused X-ray beam. The stage combination is shown in grey in Figure 1c. Pre-aligning the whole setup to the X-ray beam of the EuXFEL is done using the hexapod. For quick changes between the jet and additional reference samples or beam diagnostics, without losing the jet-to-catcher alignment, a one-dimensional stage is installed, shown in black in Figure 1c. CCD cameras with long working distance optics up to 19 cm mounted outside the vacuum on window flanges, are available at MID for observing the liquid jet and the catcher orifice. Also, a pulsed nanosecond laser can be installed to generate a shadow image of the liquid jet perpendicular to the X-ray beam for tracking jet and droplet sizes.

3. Nozzle design

3.1 Compatibility with different nozzle designs

The liquid jet setup is designed to be compatible with different nozzle types and designs. The adapter plate between the nozzle body and the Peltier-heating/cooling element is exchangeable and different nozzle holders can be mounted. Figure 1b shows an attached holder and a quartz-glass Rayleigh nozzle. Figure 2a shows an attached holder for GDVNs designed by EuXFEL [7]. This design of the holder is also compatible with other 3D-printed nozzle designs by EuXFEL. Typically, in this setup jets injected using a Rayleigh-type nozzle have a diameter in the range of 10 μm to 50 μm and for GDVNs in a range of 3 μm to 15 μm depending on the nozzle orifice size, the gas and liquid flow rates, and specifics of the sample like viscosity and surface properties.

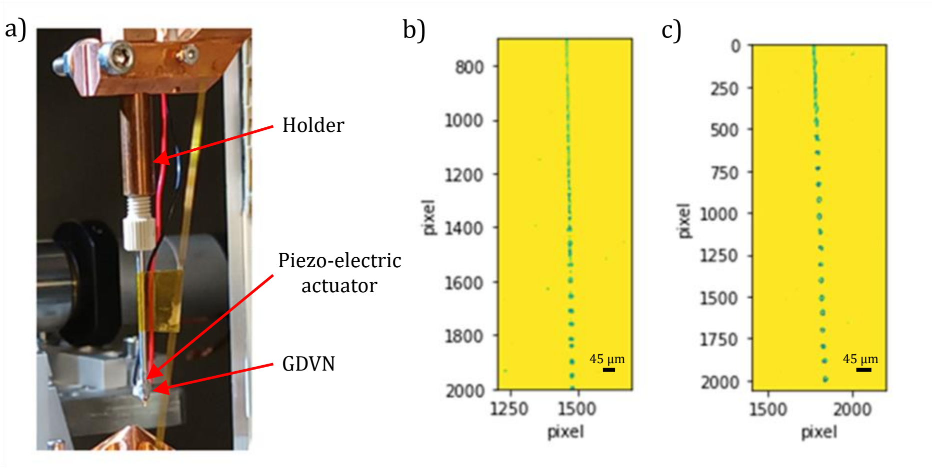

Figure 2. a) Photo of an installed 3D printed, customised GDVN. The nozzle is mounted on the temperature-controlled holder. Close to the nozzle tip a piezoelectric actuator with a resonance frequency of 450 kHz is attached. b) Shadow image of jet breakup (under ambient conditions) without piezo-electric actuator. c) Shadow image of the jet breakup (under ambient conditions) with operating piezo-electric actuator at 1511.8 kHz.

3.2 Synchronized droplet generation
To reach cold and supercooled states of liquids it is necessary to study the liquid at large distances from the nozzle orifice utilizing the effect that the sample cools rapidly because of evaporation in vacuum [22]. Due to the Rayleigh instability of the liquid jet, the liquid filament will break up into droplets and the laminar jet becomes a stream of droplets with varying shape, size and spacing between two succeeding droplets [17]. Such a break up of a jet with a diameter of ~ 9 µm and a jet speed of ~ 26 $\frac{\mathrm{m}}{\mathrm{s}}$ generated using a GDVN [7] at atmospheric pressure is shown in Figure 2b.

By attaching a piezo-electric actuator to the nozzle, the breakup can be controlled and a train of well-defined droplets is generated. Figure 2c shows the generated train of droplets of the same jet as in Figure 2b at atmospheric pressure. A burst sine voltage of 0.5 ms synchronized to the 10 Hz trigger of the X-ray beam of the EuXFEL with a voltage of 30 V and a frequency of 1511.8 kHz was applied to the piezo-electric element. The droplets spacing is about 45 µm. The EuXFEL has a high power, high repetition X-ray bunch structure that enables new types of experiments [23]. Synchronization is needed to increase the hit rate of the X-ray pulses at the interaction point with the droplet steam. For running an experiment with, e.g., 1.1 MHz repetition rate a sufficient jet speed is needed, that each X-ray pulse probes another droplet of the liquid jet [24]. After interaction with the X-ray pulses the droplet will explode and may induce a perturbation of the next droplets similar to a shock wave in the jet filament [4,24,25]. This suggests probing only every second or third droplet to obtain an unperturbed state of the sample. Assuming a droplet spacing of 45 µm this leads to a desired jet speed of approximately 100 $\frac{\mathrm{m}}{\mathrm{s}}$ to probe every second droplet with a pulse repetition rate of 1.1 MHz. The development of synchronized droplet delivery is currently under development and will be available soon.

4. Application

A demonstration scattering pattern of an X-ray diffraction experiment on water that used this system at EuXFEL is shown in Figure 3 [26]. Using a custom-made quartz-glass Rayleigh-type

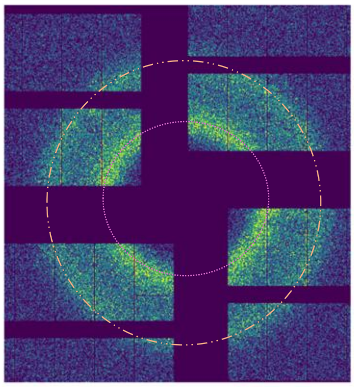

Figure 3. Zoomed in scattering pattern of a liquid water jet at a distance of 3.9 mm from the nozzle tip that was recorded with AGIPD and an X-ray beam of 23 keV in the LFOV configuration at the MID end station. The shown pattern has not been corrected for detector modules tilt, leading to the features' elliptical nature [27]. The first maximum is located at ~ 2 Å^{-1} (purple dotted line) and the second maximum at ~ 3.1 Å^{-1} (orange dashed-dotted line). The estimated liquid temperature at this distance is about 260 K [26].

nozzle with an orifice diameter of 11 μm as depicted in Figure 1 a liquid jet of ultrapure water (18.2 MΩ · cm) was injected into the evacuated multi-purpose vacuum chamber. In this experiment, the jet was scanned between ~ 3 mm to ~ 45 mm from the nozzle tip. X-ray pulses with a photon energy of 23 keV were focused to a beam size of about 5 μm FWHM on the water jet. The AGIPD detector was placed in the LFOV configuration with a sample-detector distance of about 17 cm, covering a q-range from 0 Å^{-1} to 9 Å^{-1}. Figure 3 shows the central section of a single pulse scattering pattern zoomed in on the first and second structure factor features of liquid water recorded using AGIPD, demonstrating the feasibility of such scattering experiments at MID.

5. Summary

The implementation of an experimental setup for in-air and in-vacuum experiments on liquid jets at the MID endstation of EuXFEL has been shown. The components of the nozzle holder and the catcher orifice size are compatible with different nozzle designs, including 3D printed nozzles available at EuXFEL. The setup was used successfully to study the structure and dynamics of aqueous systems in several experiments [26,28]. As a next step, a setup for piezo-electric actuator-induced breakup of the liquid jet filament into small droplet trains is under development and will become available soon.

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